Benchmarking FLUKA, PHITS, MCNPX and MARS15 Monte Carlo codes with product yields of ²⁰⁹Bi(p, x) reactions

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1. Introduction

Aiming for provision of report on shielding analysis and radiation safety issue of large accelerator facilities which have been constructing in Korea, the Monte Carlo codes, FLUKA [1], PHITS [2], MCNPX [3], and MARS15 [4], were benchmarked with comparisons to measured product yields of proton-induced reactions on Bi from 40 MeV to 100 MeV energy range which include ${}^{209}\text{Bi}(\text{p}, \text{xn}){}^{207,206,205,204,203}\text{Po}$ and ${}^{209}\text{Bi}(\text{p}, \text{m}){}^{207,206,205,204,203}\text{Po}$ pxn)^{207,206,205,204}Bi reactions. Bismuth is important as a shield material at accelerator facilities and is widely used in different nuclear technologies as a target material. To our knowledge, no systematic comparison have been performed between measured product yields of ²⁰⁹Bi(p, x) reactions and results of multiple Monte Carlo codes. However, several studies including experimentally and theoretically have been reported on the yields of radionuclei produced in Bi by a wide range of proton energies. Currently there are several Monte Carlo simulation codes that are used for activation calculation and shielding analysis of large accelerators such as PHITS, FLUKA, MCNPX, MARS15, and GEANT4. Many efforts have been made to validate and verify the physics models, which are implemented in these Monte Carlo codes.

In this work, we extend our earlier work [5] on assessment of Monte Carlo codes to predict product yields of proton-induced reactions on High-Z materials, Pb and Bi. We present the comprehensive investigation on comparison of measured product yields of protoninduced reaction on Bi, with the results calculated by using the FLUKA, PHITS, MCNPX, and MARS15 Monte Carlo codes.

2. Methods and Results

2.1 Target, Irradiation and Measurement

Two experiment rounds were carried out. Targets were prepared using the stacked-foil technique. The targets were arranged in a stack, consisting of Bi (50 μ m) activation foils, an Au (30 μ m) foil, Al (100 μ m) together with thick Pb plates. Natural Pb plates were placed between Bi activation foils to degrade proton beam energy. The typical thickness of Pb plate degrader ranged from 0.5 up to 9 mm. In each experiment, five Bi foils were used as the activation foils. Au and Al foil were used as the monitor foils. The cross-sectional size of the targets was about 5 cm × 5 cm and the total thickness of irradiated targets for the 100 MeV and 69 MeV proton beams were designed to be 1.85 and 0.91 *cm* respectively which is larger than the range of proton beams in the targets.

Irradiations took place at Korea Multi-purpose Accelerator Complex (KOMAC). The two stacked foils were irradiated for 120 and 25 seconds with 100 MeV and 69 MeV protons, respectively. The beam shape was described by Gaussian distribution at the target in both x and y coordinates based on the beam profile on the Gafchromic film.

After each irradiation, the γ -ray spectra of Bi activation foils were measured using HPGe detectors with a relative efficiency of 15 or 20 %. Regarding the halflife of a radionuclide, the gamma spectrum analysis was performed several times. The spectrum analysis was done by the Canberra's Genie-2000 gamma analysis software package (version 3.2).

2.2 Determination of proton energy and intensity

In each experiment the proton beam intensity was measured by the activation analysis method. Thin Au and Al foils were placed at the beginning of the target stacks to measure proton beam intensity via the monitor reactions ²⁷Al(p, ³p1n)²⁴Na, ¹⁹⁷Au(p, ^p1n)¹⁹⁶Au, and ¹⁹⁷Au(p, p3n)¹⁹⁴Au. The energy of incident proton beam was calibrated to be 100±0.1 MeV and 69±0.2 using multi-energy degrading techniques. The average value of obtained beam intensity using monitor reactions was used in the Monte Carlo codes to calculate product yields of residual nuclei from irradiation of thin Bi target with protons. Uncertainty of the proton beam intensity measurement by the activation analysis was estimated by possible uncertainties of measured yields $(\sim 2.0 \%)$, used cross-section $(\sim 10.0\%)$ and mass of the monitor activation foils (~0.01%). The overall uncertainty in the beam intensity measurement was estimated to be approximately 10.2%.

The energy distributions of proton beam onto the front surface of Bi activation foils were calculated by FLUKA code. According to simulation, the incident energies of protons impinging each bismuth foil were 67.5 ± 0.2 , 61.4 ± 0.7 , 54.7 ± 1.0 , 47.5 ± 1.5 , 43.1 ± 1.3 MeV, for the 69 MeV protons beam and 100 ± 0.5 , 91.8 ± 1.0 , 82 ± 1 , 72 ± 2.0 and 62 ± 2.5 MeV for 100 MeV protons beam experiment, respectively.

2.3 Monte Carlo transport codes

Monte Carlo simulations were performed by FLUKA (version 2011.2x), PHITS (version 3.02) with an inventory code, DCHAIN-SP (version 2001), MARS15 (version 2014) and MCNPX codes (version 2.7.0) with an inventory code, FISPACT (version 2010). The measured and calculated product yields of residual nuclides were compared at the end of irradiations.

2.4 Measured and calculated product yields

The results of the comparisons between the measured production yields and the calculated at different energy ranges for both 100 MeV and 69 MeV protons on Bi target are shown in Figs. 1 - 2 together with the ratios of the calculated to the measured (C/M) product yields.

2.4.1 ²⁰⁹Bi(p, 3n) ²⁰⁷Po reactions

Product yields of ²⁰⁷Po ($T_{1/2}$ = 5.80 h, ϵ : 99.98% and α : 0.02%) were obtained using the 742.72 keV ad 992.3 keV gamma-rays that were well-defined in the spectrum of each bismuth irradiated foil. The measured activities included the contribution of the decay of the simultaneously produced short-lived isomeric state ($T_{1/2}$ = 2.79 s, IT: 100%) of 207 Po. Fig. 1 Shows the measured product yields of ²⁰⁹Bi(p, 3n)²⁰⁷Po reaction together with the results of simulations. Measured product yields are in good agreement with the product yields calculated by MARS15 over the energy range of 100 to 54.7 MeV. FLUKA underestimate between factors of while, MCNPX/FISPACT 0.4 and 1. and PHITS/DCHAIN-SP overestimate the measurement by within a factor of 1.5 except for the energies lower than 50 MeV. All Monte Carlo codes show lower results than the measurements at energy less than 50 MeV.

2.4.2 ²⁰⁹Bi(p, 4n) ²⁰⁶Po reaction

The radio-nuclide ²⁰⁶Po ($T_{1/2} = 8.8$ d, ϵ : 94.55% and α : 5.45%) has a relatively long half-life. The activities were measured after few days of cooling time via 286.4, 522.47 and 807.38 keV gamma-rays. Results of simulation are close to the measurement at the energy range of 50 to 100 MeV (Fig. 2) by relatively underestimation of the measured product yields. Discrepancies between calculated and measured product yields become larger at energy less than 50 MeV.

3. Conclusions

Product yields of ²⁰⁷Po and ²⁰⁶Po radionuclei were measured using gamma-ray spectroscopy of the Bi activation foils. Measured yields were compared with calculated yields by Monte Carlo codes. FLUKA, MARS15 and PHITS tend toward underestimation of product yields of Po radionuclei over whole energy range, while, MCNPX shows both overestimation and underestimation in product yields. Results show that the predictive power of the codes has proven to be satisfactory for the 209 Bi(p, xn) reactions at energy about higher than 50 MeV.



Fig. 1. Product yield of ²⁰⁹Bi(p,3n)²⁰⁷Po reaction



Fig. 2. Product yield of ²⁰⁹Bi(p,4n)²⁰⁶Po reaction

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